Strong carrier-scattering in iron-pnictide superconductors with highest T_c obtained from charge transport experiments

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Characteristic normal-state charge transport is found in the oxygen-deficient iron-arsenides LnFeAsO$_{1-y}$ (Ln: La and Nd) with the highest T_c's among known Fe-based superconductors. The effect of "doping" in this system is mainly on the carrier scattering, quite distinct from that in high-T_c cuprates. In the superconducting regime of the La system with maximum $T_\mathrm{c} = 28$ K, the low-temperature resistivity is dominated by a T^2 term. On the other hand, in the Nd system with T_c higher than 40 K, the carriers are subject to stronger scattering showing T-linear resistivity and small magnetoresistance. Such strong scattering appears crucial for high-T_c superconductivity in the iron-based system.

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I. INTRODUCTION

Among various classes of Fe-based superconductors, the highest superconducting transition temperature T_c exceeding 50 K is realized exclusively in the LnFeAsO(F) system (Ln: lanthanide elements).[1–5] It is empirically known that T_c of LnFeAsO correlates strongly with the crystal structure and becomes maximum when FeAs$_4$ forms a nearly ideal tetrahedron, namely, when the bond angle of As-Fe-As, α, is $\sim 109.5°$.[6] However, it is still an unresolved puzzle why T_c is so different among different classes. Even in LnFeAsO(F), the optimal T_c is 28 K for Ln = La, whereas for Ln = Nd, Sm, Gd, Tb, and Dy, the maximum T_c is higher than 50 K.[7] To understand this difference in T_c, it is desirable to search for its origin in the electronic structure, as well as in the electron dynamics.

Recall that in the conventional BCS superconductors, T_c is more or less connected to scattering of the carriers in the normal-state charge transport, since the electron–phonon interaction is the origin of both the electron pairing that determines T_c and the carrier scattering that determines the resistivity.[8] For the high-T_c cuprate superconductors, the carrier scattering rate is basically proportional to temperature (T), and the T-linear scattering rate is nearly independent of material and doping. This "universal" charge transport is considered to be a hallmark of the high-T_c cuprates. As T_c is strongly material dependent, it is not directly correlated with the scattering.

Here, we investigate the transport properties of the oxygen-deficient oxypnictides LaFeAsO$_{1-y}$ and NdFeAsO$_{1-y}$ to explore how the charge transport in the normal state is correlated with the superconducting T_c in this class of pnictides, as well as in other classes of FeAs-based superconductors for comparison. Recently, single

crystals of doped BaFe$_2$As$_2$ systems have been synthesized, and their transport properties have been investigated.[9–11] Since single crystals of the LnFeAsO system are not available for a systematic study, we measured polycrystalline samples. To the best of our knowledge, this is the first systematic study of oxygen-deficient LnFeAsO$_{1-y}$, covering a wide range of doping levels and revealing the evolution of the transport coefficients, resistivity, Hall coefficient, and magnetoresistance. Since the oxypnictides are known as a multiband system with a Fermi surface consisting of three hole and two electron sheets, we have to take this into account in analyzing the transport properties by measuring the temperature and magnetic-field dependences of these transport coefficients. In this paper, we will show that T_c seems to be correlated with the scattering in the case of Fe-based superconductors.

II. EXPERIMENTAL METHODS

Polycrystalline samples of LnFeAsO$_{1-y}$ were prepared by a high-pressure synthesis technique using a cubic-anvil-type apparatus (Riken CAP-07).[12] Well-sintered samples of high density, sufficient for transport measurements, were obtained by applying a pressure of 2 GPa during the synthesis. We found no difference in the sample quality between Ln = La and Nd. Because some of the starting materials are quite reactive to oxygen, we could not prevent their oxidation in the synthesis procedure. The actual amount of oxygen deficiency y was estimated with reference to the lattice constants[6] which were slightly smaller than those of the nominal composition. There was no trace of impurity phases in x-ray diffraction patterns. In order to make the distribution of the oxygen deficiency homogeneous, the samples were annealed in air at 450 °C. The introduction of oxygen de-

ficiency is considered to work as electron doping, which seems equivalent to the substitution of fluorine for oxygen.[6,7] A standard four-terminal method was used for the resistivity measurements. Since the anisotropy ratio ρ_c/ρ_{ab} is estimated to be $\sim$25 or larger,[13,14] it is reasonable to assume that the transport properties of the polycrystalline samples mainly reflect the in-plane ones. From comparison of the transport data of polycrystalline samples and those of single crystals of other Fe-pnictide systems, such as Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$,[9,10] and also from the result for single crystalline PrFeAsO$_{1-y}$,[14] in these dense polycrystalline samples obtained by high-pressure synthesis, the grain boundary contributions to both resistivity and Hall coefficient are small, scarcely affecting their T- and H-dependences. The Hall resistivity and the transverse MR were measured in a cryostat equipped with a superconducting magnet (PPMS, Quantum Design Inc.) with the magnetic field normal to the widest plane.

III. RESULTS AND DISCUSSIONS

For the La system, superconductivity appears around $y \sim 0.08$ ("underdoped" regime), T_c becomes maximum (28 K) around $y \sim 0.11$ ("optimally" doped), and T_c decreases slowly for $y > 0.11$ in the "overdoped" regime. We name each doping regime analogously to the convention used for high-T_c cuprate superconductors for convenience. For the Nd system, T_c jumps up to above 40 K at $y \sim 0.11$, and then T_c increased gradually and reaches 52 K at $y = 0.17$, the highest y we prepared.

A. LaFeAsO$_{1-y}$

Figures 1(a) and (b) show the temperature dependence of resistivity $\rho(T)$ and the Hall coefficient $R_H(T)$ for LaFeAsO$_{1-y}$ ($0 \leq y \leq 0.23$), respectively. $R_H(T)$ was determined from the slope of the Hall resistivity ρ_H in the low-field limit, i.e. $R_H(T) = d\rho_H/dH$ at $H = 0$. $\rho(T)$ of the parent compound ($y = 0$) is only weakly T-dependent above 150 K, and it sharply drops upon the antiferromagnetic transition at $T_N \sim 150$ K and shows quadratic T-dependence with fairly large residual resistivity at the lowest temperatures. Because the antiferromagnetic order is collinear or stripe-like, the real crystal would consist of domains with different orientations, and scattering at domain boundaries would be a source of residual resistivity. The magnitude of $R_H(T)$ rapidly increases below T_N. The magnitude is larger than that above T_N by more than an order of magnitude, and R_H was ~ -0.2 cm^{3}C^{-1} at low temperatures, corresponding to the nominal electron carrier density of 3×10^{19} cm^{-3}. At low temperatures, R_H is weakly dependent on H, suggesting a contribution of at least two types of carriers, possibly two types of electrons, to R_H. The transverse magnetoresistance (MR) with the direction of H
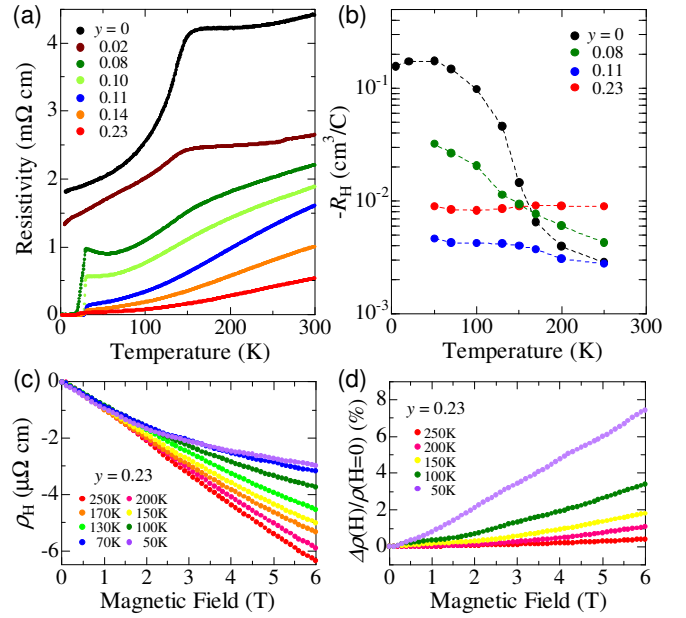

FIG. 1: (color online) (a) Temperature dependence of the resistivity of LaFeAsO$_{1-y}$ for y in the range $0 \leq y \leq 0.23$. (b) Temperature variation of the Hall coefficient R_H at zero-field limit for four representative samples with $y = 0$, 0.08, 0.11 and 0.23. Magnetic field dependence of the Hall resistivity $\rho_H(H)$ (c) and the transverse magnetoresistance (d) for $y = 0.23$.

perpendicular to that of current, $\Delta\rho(H)/\rho(0)$ ($= [\rho(H) - \rho(0)]/\rho(0)$), is quadratic in H and increases with decreasing temperature. At 5 K it reaches about 8% at $H = 6$ T. These results indicate that upon the antiferromagnetic/structural transition, the Fermi surfaces are reconstructed, and the dominant charge carriers continue to be electrons with low carrier density and relatively high mobility.

With increasing y, the magnitude of resistivity gradually decreases. Unlike the cuprates, the doping into LaFeAsO does not simply increase the carrier density, as inferred from the nonmonotonic variations of the Hall coefficient with T and y displayed in Fig. 1(b). A remnant feature of the magnetic/structural transition in $\rho(T)$ for $y = 0$ seems to remain at least up to $y = 0.10$, as shown in Fig. 1(a). This feature is suggestive of the presence of short-range magnetic/structural order which appears to coexist with superconductivity for $y = 0.08$ and 0.10, and to be a possible source of the large residual resistivity observed for all the compounds with y lower than 0.10. Also, in conjunction with the "poor" metallic behavior, the transverse magnetoresistance becomes small over the whole temperature range. Note that $\rho(T)$ exhibits a nearly parallel downshift on going from $y = 0.02$ to 0.10. This indicates that the dominant effect of doping in this regime is to reduce the residual (elastic scattering) component of resistivity (Fig. 2(b)) by suppressing the short-range order. We realize that a rapid decrease of the residual resistivity upon doping into the
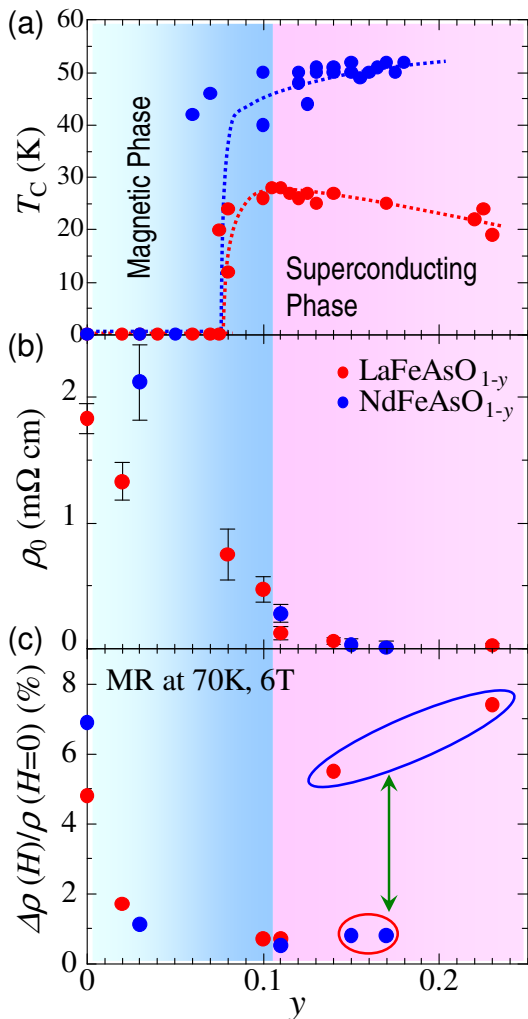
FIG. 2: (color online) (a) Critical temperatures (T_c) plotted as a function of oxygen deficiency y for LaFeAsO$_{1-y}$ (red circles) and for NdFeAsO$_{1-y}$ (blue circles). T_c follows the y dependences reported in previous papers.[6,7] (b) Residual resistivity (ρ_0) obtained from fitting (see text) plotted as a function of y. (c) The magnitude of magnetoresistance measured at $H = 6$ T and $T = 70$ K for both systems.

dominated by a T^2 term, indicative of electron–electron scattering. Such T^2 dependence up to high temperatures is widely observed in correlated electron systems, in which electron–electron interaction dominates the scattering of the carriers.[17] The T-dependence of R_H becomes weaker. Thus, the charge transport in this regime appears to show normal (good) metallic behavior.

As y further increases from 0.11, the coefficient of the T^2 resistivity decreases. In view of the increased magnitude of R_H on going from $y = 0.11$ to 0.23, it is plausible that a decrease of the scattering rate, rather than the increase of carrier density, is responsible for the observed decrease in ρ. In fact, the magnitude of MR, a measure of carrier mobility, becomes larger with increasing y (it is 1% for $y = 0.11$ and reaches $\sim 8\%$ at $H = 6$ T and $T = 70$ K for $y = 0.23$, as plotted in Fig. 2(c)). Fig. 1(c) shows the Hall resistivity for $y = 0.23$ ($T_c \sim 19$ K). At low fields ($H < 1$ T), ρ_H is linear in H and independent of T, i.e., R_H is T-independent, whereas for $H > 1$ T, nonlinear behavior is more pronounced as the temperature is lowered. The nonlinear ρ_H is clear evidence for the presence of multiple carriers. Judging from the negative sign of R_H, the nonlinearity of ρ_H, and the enhanced magnitude of R_H with respect to that for $y = 0.11$, two types of electrons contributed to ρ_H (see APPENDIX). One of the two should have higher mobility and lower density, which would have a major contribution to R_H and be the origin of the large MR. This is in contrast to the poor-metallic charge transport in the samples with lower y's, or in the "underdoped" regime. The charge transport in the "underdoped" regime is strongly influenced by disorder (inhomogeneity) produced by remnant magnetic/structural order.

Thus, the charge transport in the LaFeAsO$_{1-y}$ system does not show monotonic evolution with y, but shows a distinct behavior in different "doping" regimes. The evolution of $\rho(T)$ with y is basically the same as that reported for LaFeAsO$_{1-x}$F$_x$,[16] although the relation $y = 2x$ expected from a simple valence count does not hold, which seems to be additional evidence showing that the "doping" is not a parameter controlling carrier density alone.

B.   NdFeAsO$_{1-y}$

Figures 3(a) and (b) display $\rho(T)$ and $R_H(T)$ for NdFeAsO$_{1-y}$ ($0 \leq y \leq 0.17$), respectively. The evolution of these transport properties with y looks qualitatively similar to that for LaFeAsO$_{1-y}$. The residual resistivity sharply diminishes, and the T-dependence of R_H becomes weaker upon entering the superconducting regime. Although T_c continues to rise up to $y = 0.17$, R_H is almost T-independent, as in the "overdoped" LaFeAsO$_{1-y}$. Similarly, the magnitude of MR rapidly decreased with increasing y (Fig. 2(c)), showing a minimum at $y = 0.11$ where ρ_0 becomes vanishingly small. However, unlike LaFeAsO$_{1-y}$, MR remains very small for $y = 0.15$

"parent" material is a quite common feature observed in almost all FeAs-based superconductors so far investigated.[9–11,15,16] The Hall coefficient in this regime is strongly T-dependent. The magnitude of R_H for $y = 0.08$ increases with lowering temperature, just like that for $y = 0$, possibly due to incomplete Fermi surface reconstruction associated with the short-range order.

When y exceeds 0.10, a remarkable reduction in the residual resistivity takes place (see Fig. 2(b)), probably as a result of disappearance of the short-range order. Since the amount of oxygen deficiency increases, the reduction of the residual resistivity is evidence that the disorder due to oxygen deficiency in the LaO blocks has a minor effect on carrier scattering. For $y \geq 0.11$, i.e., in the "optimal" and "overdoped" regimes, $\rho(T)$ becomes
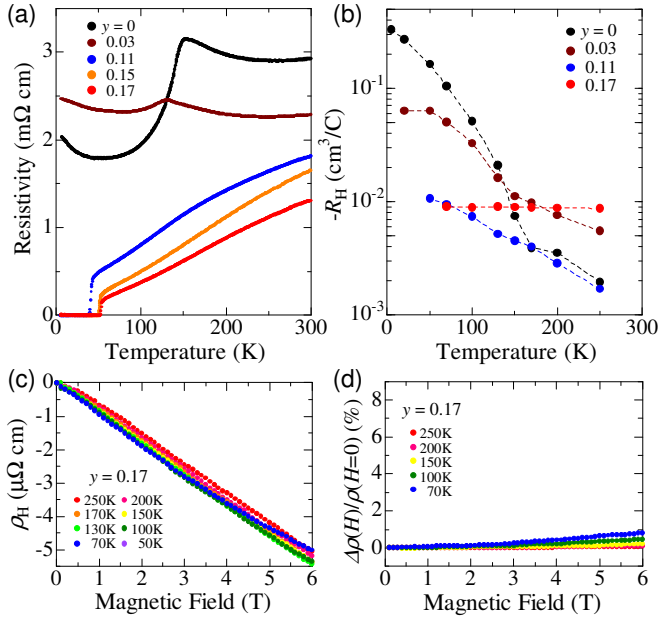
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FIG. 3: (color online) (a) Temperature dependence of the resistivity of NdFeAsO$_{1-y}$ for y in the range $0 \leq y \leq 0.17$. (b) Temperature variation of R_H at zero-field limit for the representative samples with $y = 0$, 0.03, 0.11 and 0.17. Magnetic field dependence of the Hall resistivity $\rho_H(H)$ (c) and the transverse magnetoresistance (d) for $y = 0.17$.

FIG. 4: (color online) $\log(\rho(T) - \rho_0)$ vs. $\log T$ plots for LaFeAsO$_{1-y}$, CeFeAsO$_{1-y}$, PrFeAsO$_{1-y}$ and NdFeAsO$_{1-y}$ at various doping levels, where ρ_0 is the residual component of resistivity. The and dashed lines are guides for the eye showing the slopes of $\rho(T) \sim T^n$.

and 0.17. The most remarkable difference is that the normal-state resistivity in the superconducting regime (y = 0.11, 0.15 and 0.17) is dominated by a T-linear term. The T-linear resistivity is reminiscent of optimally doped high-T_c cuprates, but the T-independent R_H is in sharp contrast with that for cuprates, in which R_H increases with decreasing T.[18]

Figures 3(c) and (d) show the magnetic-field dependences of the Hall resistivity ρ_H and the transverse MR for the $y = 0.17$ sample. ρ_H is linearly dependent on H up to 6 T and does not show an appreciable temperature dependence; that is, R_H is independent of both T and H. MR is remarkably small even at the lowest temperature (70 K), less than 1% at $H = 6$ T. This result suggests that in the $y = 0.17$ sample, only one type of electron carriers with fairly low density ($\sim 7 \times 10^{20}$ cm^{-3} or ~ 0.05 per Fe) dominates in R_H and probably in $\rho(T)$. Other carriers with perhaps comparable or higher density would be ineffective in charge transport due either to the much lower mobility or to gapping in the corresponding Fermi surface. From the small MR, it follows that even the electrons which dominate in R_H suffer strong scattering, giving rise to the T-linear resistivity.

C. T_c–n correlation

To reveal the correlation between charge transport and T_c, we investigated the exponent n of $\rho(T) \sim T^n$ for each sample. We fitted the data in the form of $\rho(T) = \rho_0 +$
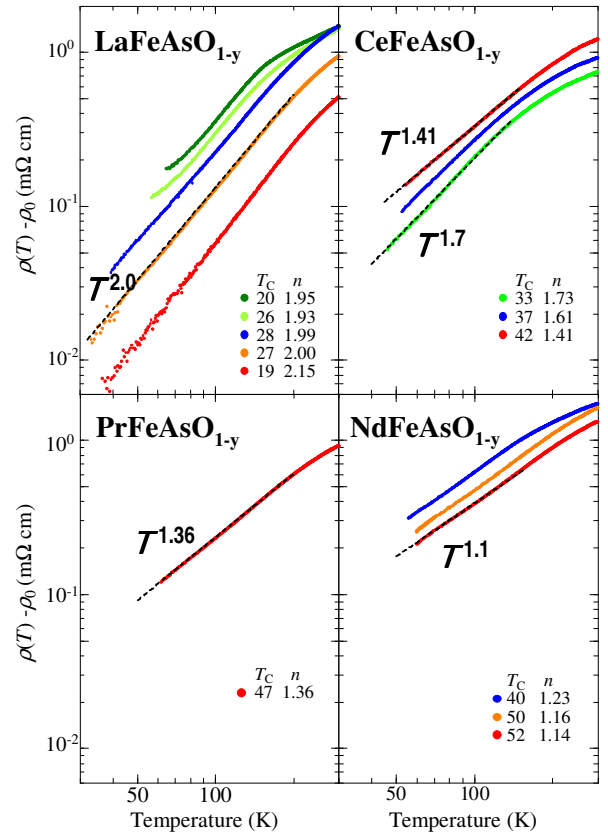
$A\,T^n$ in the temperature range between T just above T_c and $T = 150$ K (Fig. 4), where ρ_0 is the residual component of resistivity. The exponent n plotted against T_c is shown in Fig. 5. NdFeAsO$_{1-y}$ with higher T_c clearly belongs to a class distinct from LaFeAsO$_{1-y}$. On the same figure are also plotted the n-values for CeFeAsO$_{1-y}$ and single crystals of PrFeAsO$_{1-y}$ with T_c between 30 and 47 K, as well as for K-, Co- and P-substituted BaFe$_2$As$_2$. Except for Co- and P-substituted BaFe$_2$As$_2$, the data points sit in between LaFeAsO$_{1-y}$ and NdFeAsO$_{1-y}$, showing a trend that n decreases from 2 to 1 as T_c increases. Note that the compounds showing this T_c–n correlation have "dopant" or disorder sites located outside the FeAs blocks.[1,3] $\rho(T)$ of Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ and BaFe$_2$(As$_{1-x}$P$_x$)$_2$ is nearly linear in T in their highest T_c region, whereas the maximum T_c is 25 K in the former and 30 K in the latter. Disorder in the Fe-layer is expected to affect charge transport and T_c more seriously. Also, it is known that T_c is sensitive to the As-Fe-As bond angle or the pnictogen height from the Fe layer. Therefore, the P-substitution for As sites locally modulate the bond angle / pnictogen height, which would act to reduce T_c.
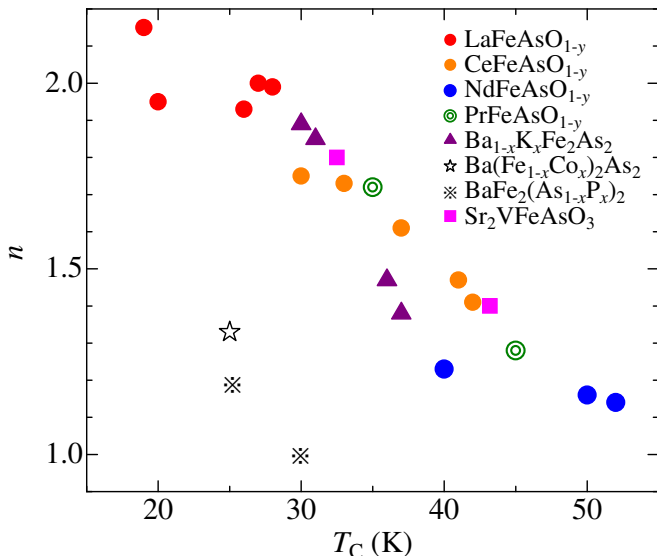
FIG. 5: (color online) Relationship between T_c and the exponent n of $\rho(T) \sim T^n$ for various kinds of FeAs-based superconductors. The closed symbols are for $LaFeAsO_{1-y}$ and $NdFeAsO_{1-y}$, and the others are for other systems[19], including single crystals of $PrFeAsO_{1-y}$.[14] The results for K- and Co- doped $BaFe_2As_2$ are in agreement with those reported by other groups. The data for P-doped $BaFe_2As_2$ and $Sr_2VFeAsO_3$ is from Ref. 11 and 21, respectively.

The magnitude of MR is always small for superconducting $NdFeAsO_{1-y}$ showing T-linear resistivity, whereas MR is much larger in $LaFeAsO_{1-y}$ with resistivity dominated by a T^2 term. Thus, one may conclude that strong "inelastic" carrier scattering is the origin of the T-linear resistivity in the normal state and a possible ingredient for achieving T_c of 40–50 K in Fe-based superconductors. Electron–phonon interaction is probably not the origin of the T-linear resistivity in the Fe-based superconductors, as it is unlikely that the electron–phonon scattering gives rise to T-linear resistivity in some cases and to T^2 resistivity in others in the same class of compounds.[20]

This correlation between the exponent n and T_c is not restricted to LnFeAsO$_{1-y}$ investigated in the present work.[22,23] In the case of LaFePO, T_c is as low as ~ 7 K, and $\rho(T)$ obeys a T^2-dependence over a wide T range.[24,25] A crossover from T^2 to T-linear dependence of resistivity associated with an increase of T_c is seen in one system, $La_{1-x}Y_xFeAsO_{1-y}$, in which T_c goes up to 43 K with x,[26] and also in $NdFeAsO_{1-y}$ under pressure where n increases from about 1 to 2 as T_c decreases with pressure.[27]

D. Comparison with theories

In the band structure of nondoped LaFeAsO,[28] the top of a hole band (called a γ band) centered at Γ is located just above the Fermi level. It moves down with electron doping and finally is located below the Fermi surface. In the case of $NdFeAsO_{1-y}$ the top of the γ band is higher than that for $LaFeAsO_{1-y}$ being always above the Fermi level regardless of electron doping level. It is argued that a Fermi surface derived from the γ band facilitates nesting between disconnected portions of electron Fermi surfaces, which would enhance "interband" scattering of electrons between the two nesting Fermi surfaces and, in certain circumstances (e.g., when coupled with spin fluctuations), would be a source of pairing interaction.[29,30] In addition, the location of the γ band is found to be sensitive to local crystal structure, specifically, the As height from the Fe plane or the bond angle α of the FeAs$_4$ octahedron.[28] Then, the γ-Fermi surface seems to bridge between the known T_c-α correlation and the present T_c-n correlation we observed. Unfortunately, there is no experimental evidence for the presence of the γ-Fermi surface in $NdFeAsO_{1-y}$ and its absence in $LaFeAsO_{1-y}$.

An alternative scenario is that $NdFeAsO_{1-y}$ showing T-linear resistivity is located near some quantum critical point (QCP), and electrons are subject to strong scattering from quantum critical fluctuations, as is discussed in the heavy fermion systems for a source of T-linear resistivity. A possible QCP in Fe-based systems is the point at which the magnetic or structural order, either long- or short-range order, disappears on the doping axis. In the present case, it would be $y \sim 0.1$, where the residual resistivity sharply drops and high-T_c superconductivity sets in. Then, a question would arise: what makes $LaFeAsO_{1-y}$ distinct from $NdFeAsO_{1-y}$ in that the T^2 term dominates in resistivity even in the vicinity of $y \sim 0.10$?

IV.   CONCLUSION

The present results strongly suggest that the strong carrier scattering which gives rise to T-linear resistivity is intimately correlated with high-T_c in the Fe-based superconductors. In conventional BCS superconductors, the electron–phonon scattering rate, which determines the normal state resistivity, is linked to the strength of the pairing interaction in the superconducting state. A correlation between T-linear resistivity and high T_c is also discussed in the context of quantum criticality in organic, heavy fermion, and cuprate superconductors, as a common feature of the normal state of unconventional superconductors.[31] However, in the case of cuprates, the carrier scattering rate is not necessarily related with T_c. T_c is controlled by the superfluid density, and the transport scattering rate is not strongly dependent on doping and material in the underdoped and optimally doped regimes. In view of the charge transport demonstrated in the present work, the Fe-based superconductors apparently belong to a class different from the cuprates, and seem to have an aspect in common with other classes of superconductors in that the carrier scattering has some relevance to T_c.
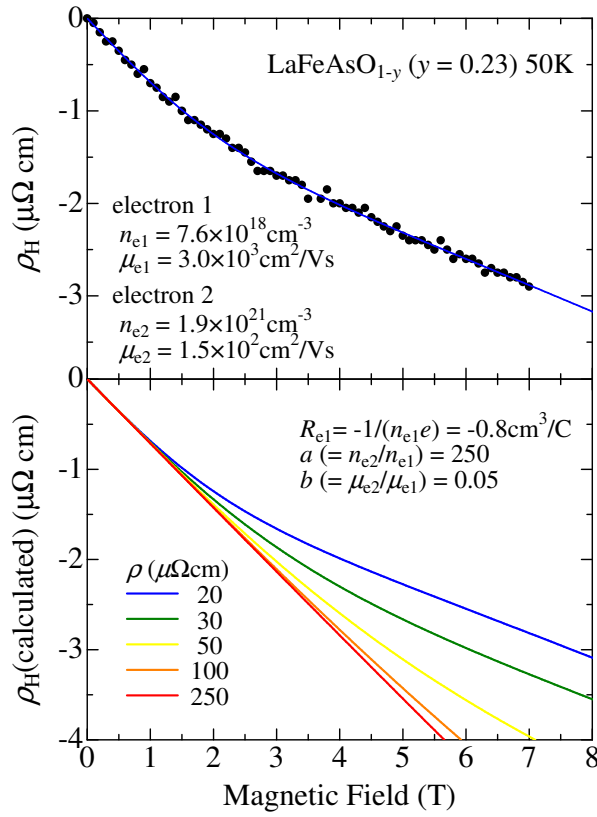
FIG. 6: (color online) (a) The magnetic field dependence of Hall resistivity ρ_H at 50 K for LaFeAsO$_{1-y}$ ($y = 0.23$). The blue line was obtained by fitting assuming a two-carrier model. (b) The calculated results for some temperatures.

APPENDIX

By fitting $\rho_H(H)$ assuming a two-carrier model, we obtained a result that two types of electron carriers reproduce the data in Fig. 1(c), as follows. Namely, $n_{e1} \sim 7.6 \times 10^{18}$ cm^{-3}, $\mu_{e1} \sim 3.0 \times 10^3$ cm^{2}V^{-1}s^{-1}, $n_{e2} \sim 1.9 \times 10^{21}$ cm^{-3}, and $\mu_{e2} \sim 1.5 \times 10^2$ cm^{2}V^{-1}s^{-1} at $T = 50$ K, where n_{ei}, μ_{ei}, and i represent carrier number, mobility, and type of carriers ($i = 1$ and 2), respectively. The temperature dependence of $\rho_H(H)$ is also reproducible, assuming the same temperature dependence of both μ_{ei}'s as that of the inverse of resistivity. Note that the existence of the minor carrier ($i = 1$) with high mobility enhances the absolute value of R_H at the low-field limit, which is larger than that simply expected for the major carrier ($i = 2$). Although the obtained parameter set is somewhat unrealistic and the origin of the minor carrier is not clear from a band calculation, the nonlinearity of $\rho_H(H)$ may be related to the configuration of Fermi surfaces.

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